Supplementary Information

Intrinsic and Extrinsic Charge Transport in $CH_3NH_3PbI_3$ Perovskites Predicted from First-Principles

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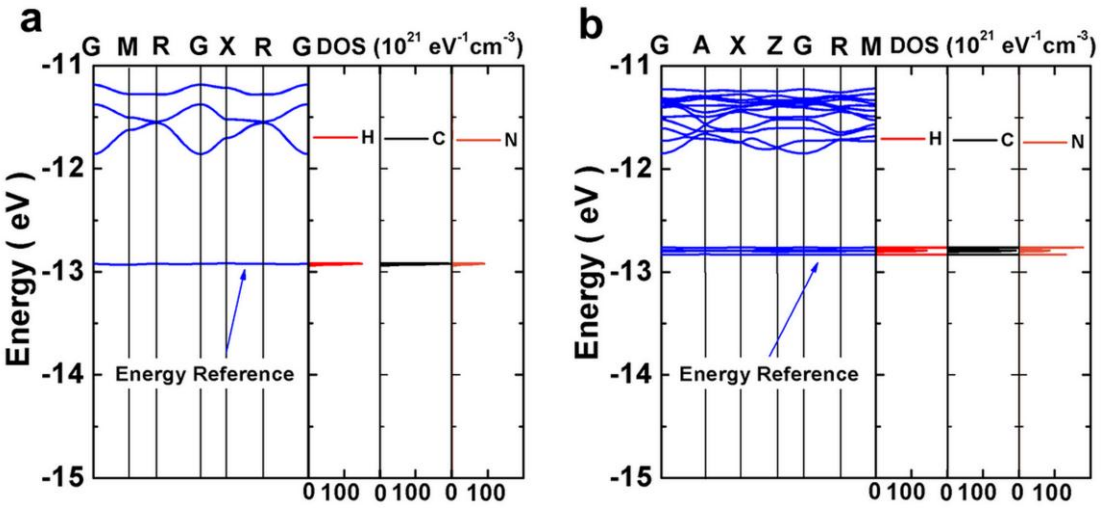
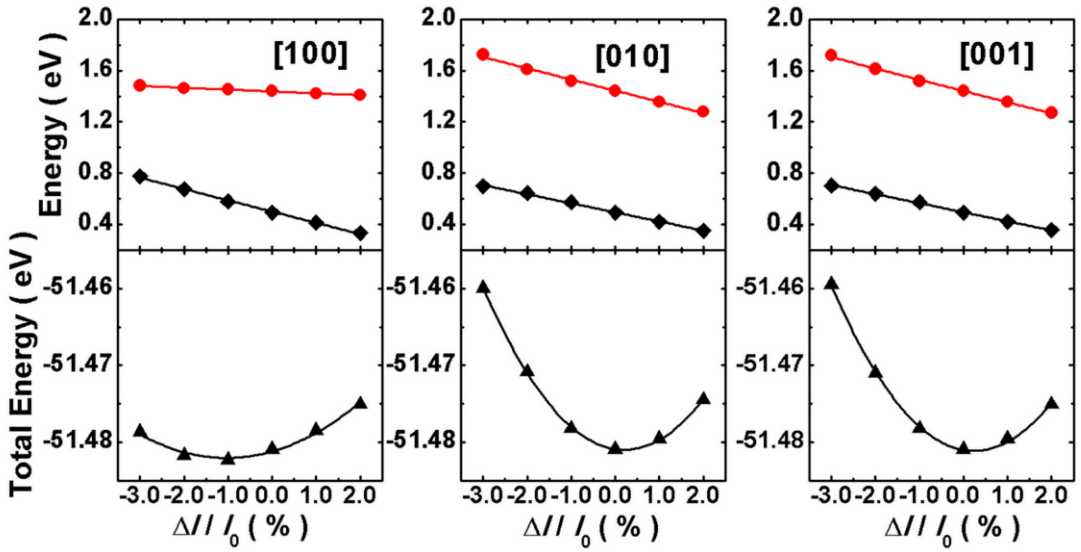
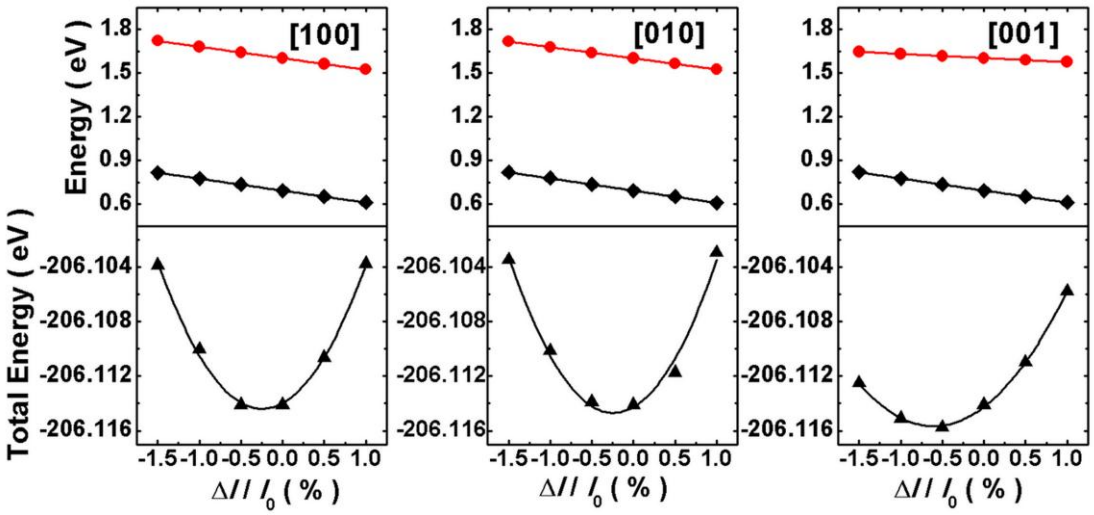


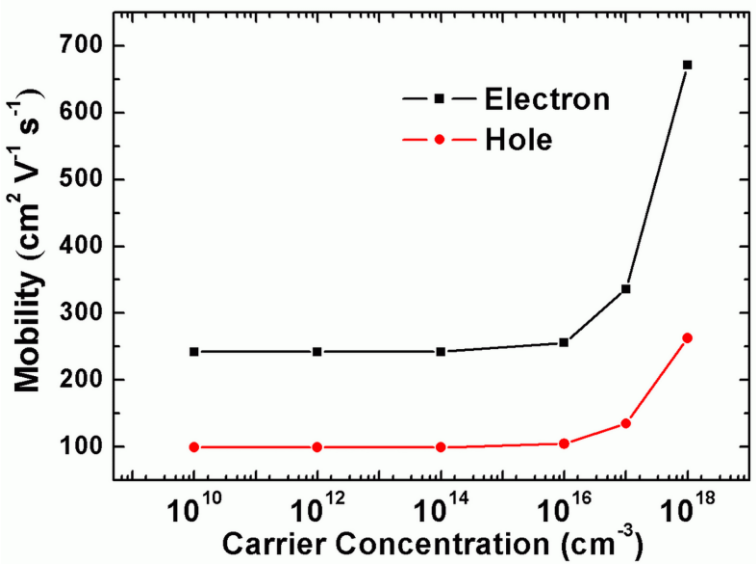
Supplementary Figure S1. Energy reference for the VBM and CBM calibration during straining of cubic (a) and tetragonal (b) $CH_3NH_3PbI_3$.



Supplementary Figure S2. Energy shift of the CBM and VBM with respect to dilation (upper panel) and total energy change with dilation (lower panel) along [100], [010] and [001] directions for cubic $CH_3NH_3PbI_3$.



Supplementary Figure S3. Energy shift of the CBM and VBM with respect to dilation (upper panel) and total energy change with dilation (lower panel) along [100], [010] and [001] directions for tetragonal $CH_3NH_3PbI_3$.



Supplementary Figure S4. Mobility of tetragonal $CH_3NH_3PbI_3$ perovskites limited by charged impurity scattering as a function of free carrier concentration at an impurity density of 10^{18} cm^{-3}.

Supplementary Table S1. Structural parameters of cubic and tetragonal $CH_3NH_3PbI_3$.

		a (Å)	*b* (Å)	*c* (Å)	α	β	γ
Cubic	Calc.	6.67	6.44	6.44	89.76	90.02	90.02
	Expt.[a]	6.31	6.31	6.32	90.0	90.0	90.0
Tetragonal	Calc.	9.11	9.07	12.91	90.25	89.25	88.32
	Expt.[a]	8.85	8.85	12.64	90.0	90.0	90.0

[a]Ref. S1.

Supplementary Table S2. Effective mass m^*/m_e of electrons and holes in cubic and tetragonal $CH_3NH_3PbI_3$. 1 and 2 represent bands with opposite spins.

		VB(2)	VB(1)	CB(1)	CB(2)
Cubic	R-M	0.60	0.28	0.16	0.30
	R-G	0.52	0.26	0.17	0.28
	R-X	0.55	0.31	0.18	0.32
Tetragonal	G-M	0.52	0.28	0.15	0.23
	G-Z	0.45	0.43	0.26	0.27
	G-X	0.31	0.24	0.16	0.20
	G-A	0.50	0.28	0.15	0.24
	G-R	0.36	0.26	0.17	0.23

Supplementary Table S3. Relaxation times of electrons and holes in cubic and tetragonal $CH_3NH_3PbI_3$ subject to impurity and acoustic phonon scatterings at 300 K. The impurity density is taken as 10^{16}, 10^{17} and 10^{18} cm^{-3} respectively. The free carrier concentration is 10^{14} cm^{-3}.

τ (ps)		Cubic		Tetragonal	
		Electron	Hole	Electron	Hole
Ionized Impurity	10^{18} cm^{-3}	0.017	0.019	0.016	0.018
	10^{17} cm^{-3}	0.17	0.19	0.16	0.18
	10^{16} cm^{-3}	1.67	1.95	1.59	1.83
Total	10^{18} cm^{-3}	0.011	0.017	0.013	0.017
	10^{17} cm^{-3}	0.042	0.10	0.065	0.13
	10^{16} cm^{-3}	0.082	0.30	0.16	0.51

Supplementary Table S4. Mobilities of electrons and holes in cubic $CH_3NH_3PbI_3$ subject to impurity and acoustic phonon scatterings at 300 K. The impurity density is taken as 10^{16}, 10^{17} and 10^{18} cm^{-3} respectively. The free carrier concentration is 10^{14} cm^{-3}.

μ (cm^2 V^{-1} s^{-1})		electron			hole		
		a	*b*	*c*	*a*	*b*	*c*
Ionized Impurity	10^{18} cm^{-3}	288	172	172	88.7	151	151
	10^{17} cm^{-3}	2878	1716	1718	887	1506	1506
	10^{16} cm^{-3}	28781	17157	17175	8871	15062	15065
Total	10^{18} cm^{-3}	164	101	101	72.2	119	119
	10^{17} cm^{-3}	473	303	303	377	592	591
	10^{16} cm^{-3}	708	476	476	927	1399	1399

Supplementary Table S5. Mobilities of electrons and holes in tetragonal $CH_3NH_3PbI_3$ subject to impurity and acoustic phonon scatterings at 300 K. The impurity density is taken as 10^{16}, 10^{17} and 10^{18} cm^{-3} respectively. The free carrier concentration is 10^{14} cm^{-3}.

μ (cm^2 V^{-1} s^{-1})		electron			hole		
		a	*b*	*c*	*a*	*b*	*c*
Ionized Impurity	10^{18} cm^{-3}	184	185	242	158	161	99.2
	10^{17} cm^{-3}	1844	1853	2423	1577	1613	992
	10^{16} cm^{-3}	18442	18534	24228	15768	16132	9918
Total	10^{18} cm^{-3}	139	139	178	142	145	90.2
	10^{17} cm^{-3}	590	585	711	930	949	605
	10^{16} cm^{-3}	1294	1261	1365	3086	3150	2050

Supplementary References

S1. Stoumpos, C. C., Malliakas, C. D. & Kanatzidis, M. G. Semiconducting tin and lead iodide perovskites with organic cations: Phase transitions, high mobilities, and near-infrared photoluminescent properties. *Inorg. Chem.* **52,** 9019-9038 (2013).